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Light Scattering Intensity Fluctuations in Microdroplets Containing Inclusions

by Gorden Videen, Paul Pellegrino, Dat Ngo,
John S. Videen, and Ronald G. Pinnick

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Gorden Videen and Paul Pellegrino
Information Science and Technology Directorate, ARL

Dat Ngo
NgoCo

John S. Videen
University of California at San Diego

Ronald G. Pinnick
Information Science and Technology Directorate, ARL

DTIC QUALITY INSPECTED 3

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Abstract

A prominent characteristic of light scattered from a microparticle containing inclusions is fluctuating intensity due to changing positions of the inclusions with respect to each other and the host droplet. We calculate the magnitude of these fluctuations for a host sphere containing a single nonconcentric spherical inclusion and experimentally measure the fluctuation amplitudes for host spheres containing multiple inclusions. We find that for relatively small single inclusions, the amplitude of the scattering fluctuations increases approximately linearly with the area of the inclusion. For multiple inclusions, the fluctuation amplitude increases with concentration, with an approximate power-law dependence.

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1. Introduction

A great deal of interest has recently developed in the elastic [1-5] and inelastic [6-11] scattering of host droplets containing small inclusions. The scattered intensity from such systems fluctuates with time, as the relative positions of the inclusions change. Such systems are of interest in atmospheric optics, since atmospheric water droplets and aerosols generally contain inhomogeneities that can significantly affect the scattering and absorption properties of the host [12]. Knowing the effects of microcontaminants on the scattering and absorption by water droplets and aerosols is important in radiative transfer calculations [13,14]. These effects are also of interest in scattering from biological systems (cells), which are generally inhomogeneous and may undergo refractive index changes with time.

The inverse problem of calculating the contaminant characteristics from the scattering signal should not be overlooked: information about the scattering system is often our primary interest. We realize the difficulties in this latter task for our system of a host particle containing small inclusions when we consider the relative sizes of the particles involved. Scattering signals of two particles in close proximity do not add linearly, especially when one of the particles is encapsulated within the other; nevertheless, we know that the scattering effect of an inclusion whose volume is orders of magnitude smaller than the host is small [15]. Furthermore, the inclusion does not add additional structure to the scattering phase function of the host; the relative amplitudes of the maxima and minima may be altered somewhat, but the number of spherical harmonics necessary to describe the host, with or without an inclusion, is approximately the same [16-20]. The time-varying intensities remain to be analyzed.

We examine the time-varying intensities in two separate experiments. First, we examine the effects of a single spherical inclusion on the scattering intensity fluctuations. We performed this investigation numerically, since it is rather difficult to prepare samples to such precision, and computer codes exist that make such an investigation routine [16-26]. Second, we examine the effects of inclusion concentration. We performed this investigation experimentally, since the number of computations currently required for an adequate numerical study make such an approach impractical.

2. Single Inclusions

We begin our analysis by examining numerical results calculated from a host sphere containing a single, nonconcentric, spherical inclusion. We choose this particular system because it is the simplest encapsulated system for which a complete theory has been derived [16–26]. Two relevant systems are examined: a glycerol host containing a polystyrene latex inclusion, and a water host containing a carbon inclusion. Figure 1a shows the standard deviation of the backscatter intensity σ_{back} (reflecting the intensity fluctuations) as a function of latex inclusion radius r_{inc} . We calculate the backscatter intensities (Mueller matrix element S_{11}) by letting the inclusion position vary systematically throughout the host. The density of inclusion positions is doubled until the results vary by less than 1 percent from those of the previous density. Since we assume that the inclusion placement within the host droplet is arbitrary, we neglect the effects of surface tension. Although neglecting these effects may appear an oversimplification, pictures of water droplets containing carbon inclusions show the inclusions to be placed “randomly throughout the volume of each drop” [12]. For at least some applications, the assumption of a random distribution is appropriate, and we use it in our calculations. It should be noted that these calculations are extremely computer intensive; for instance, the 10 data points calculated for host radius $r_{host} = 3.0 \mu\text{m}$ took approximately 10 cpu days on a Silicon Graphics 8000-series processor running at 1.4 Gflops.

Since the computer time necessary to make the calculations is approximately proportional to r_{host}^5 , we limit ourselves to calculations of relatively small host radii. Even so, certain trends are apparent in these data. First, the amplitude of the intensity fluctuations tends to increase for small inclusion radii, eventually reaching a maximum value, before dropping sharply toward zero as the inclusion fills the entire volume of the host sphere. Second, the intensity fluctuations appear to have little dependence on the host radius. Increasing the glycerol host radius by 50 percent has only a minor effect on the fluctuation amplitudes. The fluctuation amplitudes must show a dependence on the host refractive index m_{host} , because when m_{host} approaches the refractive index of the inclusion or the incident medium, the system reduces to an isolated Mie sphere for which there are no intensity fluctuations. This could be

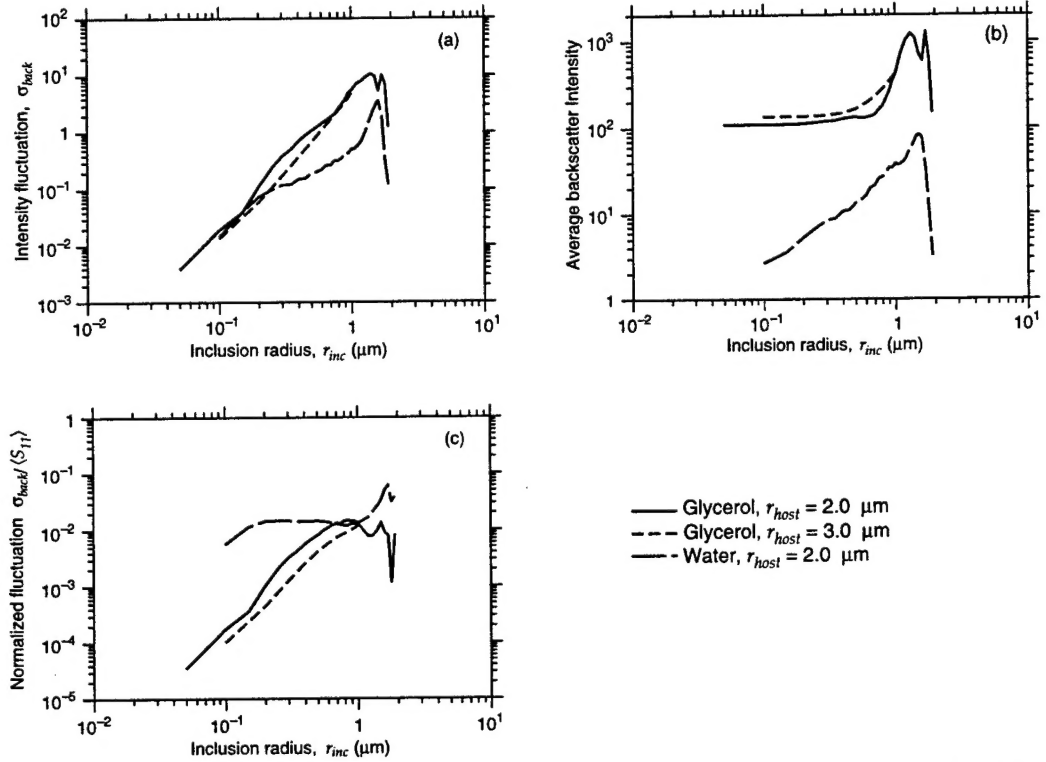


Figure 1. Comparison of scattering parameters of a spherical host containing single spherical inclusion as a function of inclusion radius ($\lambda = 647.1 \text{ nm}$): (a) calculated amplitude of intensity fluctuations, (b) average backscatter intensity, and (c) normalized intensity fluctuation amplitudes (intensity fluctuation amplitude normalized to average backscatter intensity). Data points are calculated for glycerol hosts ($m_{\text{host}} = 1.471$) containing latex inclusions ($m_{\text{inc}} = 1.59$) and for water hosts ($m_{\text{host}} = 1.335$) containing carbon inclusions ($m_{\text{inc}} = 1.94 + 0.66i$).

why the water host, having a lower refractive index than the glycerol host, displays a lower level of intensity fluctuations. Third, when the inclusion radius is relatively small compared with the host radius, the amplitude of the intensity fluctuations appears to have a power-law dependence—that is, an approximately quadratic dependence on the inclusion radius, increasing approximately linearly with the area of the inclusion.

Figure 1b shows the average backscatter intensity (over all inclusion positions within the host) as a function of the inclusion radius for the scattering systems shown in figure 1a. As the inclusion radius is increased, the average backscatter intensity also increases before decreasing to previous levels. The shape of these curves can be attributed to enhanced backscatter (EBS), which has been studied extensively from irregular surfaces [27–32]. EBS has been attributed to constructive interference of rays reflecting off multiple interfaces. The path difference is the same when the order of the interfaces that the light ray strikes is reversed for backscattered light; therefore, the forward and backward traversing rays interfere constructively, and the resulting intensity is enhanced. As the inclusion size increases, the inclusion scatters more light, contributing to this effect. As its size approaches that of the host sphere, the phase differences acquired by rays traveling through the host sphere become negligible, and the EBS disappears.

Figure 1c is the normalized fluctuation intensity: i.e., the standard deviation of the backscatter intensity, shown in figure 1a, divided by the average backscatter intensity shown in figure 1b. This quantity corresponds to the proportion of intensity fluctuation or “noise” on the average signal, and is readily measurable. The normalized fluctuation intensity is extremely sensitive to system parameters. For the water host, the fluctuations are relatively constant, remaining approximately 1 percent of the average backscatter intensity signal, as the carbon inclusion size is increased from the Rayleigh regime until it is nearly the size of the host sphere. For the glycerol hosts, the amplitude of the intensity fluctuations again has an approximate power-law dependence on the inclusion radius, and for these parameters, the power-law exponent is approximately 2; i.e., the normalized intensity fluctuations are approximately proportional to the area of the inclusion.

3. Multiple Inclusions

In many practical instances, like droplet seeding, the host droplet contains more than one inclusion. It is therefore desirable to examine the effects of inclusion concentration on the intensity fluctuations. Although theories have been derived to calculate the scatter from a host containing multiple inclusions [18,19,22], it is currently impractical to make intensity fluctuation calculations for these types of scattering systems because of the enormous number of computations required. We therefore examine experimentally the fluctuation dependence on concentration; however, these experimental results are not meant to be compared with the theoretical results of the previous section. The results of this section are meant to provide additional information on the behavior of the scattered intensities as a function of the inclusion parameters: in this case, the inclusion concentration.

In our experiment, a charged glycerol droplet having radius $r_{host} \sim 10$ to $12 \mu\text{m}$ is captured in an electrodynamic trap [33]. The droplet is generated with a spray atomizer prepared to contain a known concentration of uniform spherical latex inclusions having nominal radius r_{inc} . The solution contains a small amount of surfactant to prevent clumping. We determined the host droplet size to within 5 percent by measuring the positions of the first two minima of the Fraunhofer pattern using the technique outlined by Chen [34]. Levitated droplets are illuminated with a krypton-argon laser beam ($\lambda = 647.1 \text{ nm}$), and the scattered light is detected by photomultiplier tubes placed in the forward-scatter direction ($\sim 7^\circ$) and in the backward-scatter direction ($\sim 179.5^\circ$). The detectors collect light over a relatively small conical solid angle (approximately 0.1° half angle). The signals are amplified, fed through a low-pass filter (250 Hz) to remove detector shot noise, and digitized with an 820-Hz analog-to-digital recorder. To minimize evaporation, we hold the droplets for approximately 1 hour in the electrodynamic trap, so that equilibrium with the vapor within the containment vessel enclosing the trap can be attained. However, since the containment vessel is not completely airtight (it contains holes to allow for entrance and exit of the laser beam), some vapor does escape and the host droplet slowly evaporates. The rate of evaporation is approximately 0.2 nm over a 6-s data run.

In order to estimate relative errors in the intensity fluctuations, we produce 3 to 5 droplets for each concentration and measure the intensities over 20 data runs for each droplet. Since we do not know the exact number of inclusions in each droplet, only the inclusion concentrations in solution, we examined several droplets at each concentration to find their average scattering characteristics. Figure 2 shows the average of the standard deviations of the experimental forward-scattered intensities, normalized to the average scattered intensity as a function of latex inclusion concentration, along with their relative uncertainties for two different-size latex inclusions ($r_{inc} = 0.25, 0.5 \mu\text{m}$). As the inclusion concentration increases, the amplitude of the intensity fluctuations also increases. The amplitude of the intensity fluctuations is much larger for the hosts containing the larger inclusions, and remains larger even when the inclusion concentration is expressed as a volume fraction. Figure 2 also shows regression curves for each inclusion size. The amplitude of the intensity fluctuations increases with inclusion concentration following a power law; the power-law exponent is dependent on the inclusion size.

For the system of host droplets containing multiple inclusions, the backscatter intensity fluctuations vary dramatically with droplet size as well as inclusion concentration. These variations are so large that a clear dependence

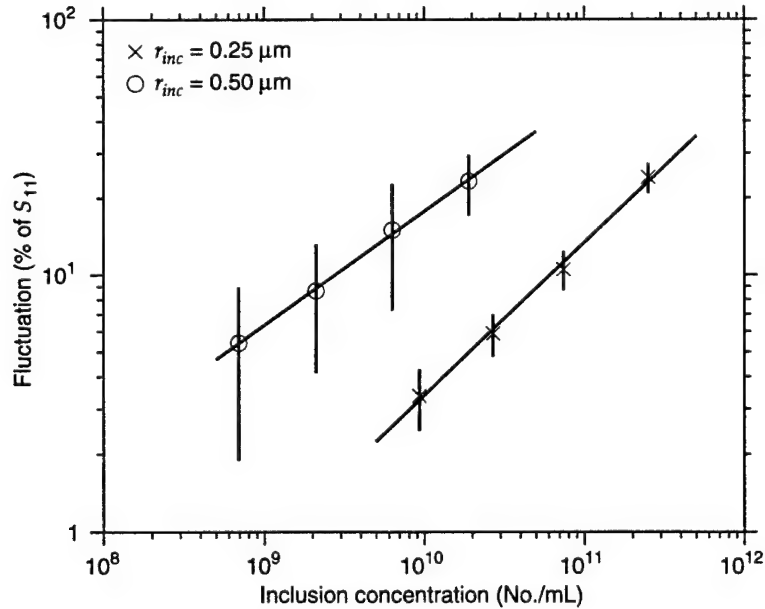


Figure 2. Experimental intensity fluctuation amplitudes from $r_{host} \sim 10$ to $12 \mu\text{m}$ glycerol hosts as a function of latex inclusion concentration. Number of inclusion spheres in $r_{host} \sim 10 \mu\text{m}$ host is approximately 4.2×10^{-9} ml times concentration.

of backscatter intensity fluctuation with concentration is not evident in our data. It is much easier to see a trend in the intensity fluctuation as a function of concentration if we look at the scatter in the forward direction. For smaller concentrations of smaller inclusions, we would expect the backward-scatter intensities to provide useful information, as demonstrated by the theoretical results shown in figure 1.

4. Results

We have explored the dependence of the intensity fluctuations found in microdroplets containing inclusions. We found that for microdroplets containing a single, relatively small spherical inclusion, the amplitude of the intensity fluctuations increases approximately linearly with the area of the inclusion. Experimentally, we found that as the inclusion concentration increases, so does the amplitude of the fluctuations. The rate at which the intensity fluctuations increase is dependent on the inclusion size and also appears to have a power-law dependence on concentration.

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